Does Fog Chemistry in Switzerland Change With Altitude?

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Abstract

During two extended summer seasons in 2006 and 2007 we operated two battery driven versions of the Caltech active strand cloud water collector (MiniCASCC) at the Niesen mountain (2362 m a.s.l.) in the northern part of the Swiss Alps, and two devices at the Lägeren research tower (690 m a.s.l.) at the northern boundary of the Swiss Plateau. During these two field operation phases we gained weekly samples of fog water, where we analyzed the major anions and cations, and the isotope ratios of fog water (in form of δ^2H and δ^{18}O). Dominant ions in fog water at all sites were NH_4^+, NO_3^-, and SO_4^{2-}. Compared to precipitation, the enrichment factors in fog water were in the range 5–9 at the highest site Niesen Kulm. We found considerably lower summertime ion loadings in fog water at the two Alpine sites than at lower elevations above the Swiss Plateau. Lowest ion concentrations were found at the Niesen Kulm site at 2300 m a.s.l., whereas highest concentration (a factor 7 compared to Niesen Kulm) were found in fog water at the Lägeren site. Occult nitrogen deposition was estimated from fog frequency and typical fog water flux rates. This pathway contributes 0.3–3.9 kg N ha^{-1} yr^{-1} to total N deposition at the highest site on Niesen mountain, and 0.1–2.2 kg N ha^{-1} yr^{-1} at the lower site. These inputs are the reverse of ion concentrations measured in fog due to the 2.5 times higher frequency of fog occurrence at the mountain top (overall fog occurrence was 25% of the time) as compared to the lower Niesen Schwandegg site. Although fog water concentrations were on the lower range reported in earlier studies, fog water is likely to be an important N source for Northern Alpine ecosystems and might reach values up to 16% of total N deposition and up to 75% of wet N deposition by precipitation.

Keywords: Fog chemistry, Cloud chemistry, Swiss Alps, Mountain fog, Stable isotopes in fog water

1. Introduction

Fog and cloud water can be a significant source of water input to montane ecosystems [Walmsley et al., 1996; Dawson, 1998; Bruijnzeel et al., 2005], and chemical solutes in fog and cloud water can add relevant amounts of nutrients or pollutants to these ecosystems (e.g., Thalmann et al. 2002; Burkard et al. 2003). Although in Central Europe fog does not yield significant amounts of water compared to precipitation, deposition of nutrients such as nitrogen or sulphur can lead to over-nutrition, mainly in forest ecosystems [Thalmann et al., 2002]. However, it is unknown, whether such findings also apply to mountain peaks, or only to lower mountains that are within the anthropogenically polluted atmosphere. We addressed this question by collecting fog water and analysing its composition at four sites grouped at two localities, (a) Mount Niesen, Switzerland (2362 m a.s.l.), and (b) the low mountain site Lägeren (682 m a.s.l.) (Figure 1). Many factors influence the chemical composition of fog and cloud water, namely the provenience of the air mass, cloud dynamics, and microphysics [Cini et al., 2002]. In industrialized countries, concentrations of solutes are generally higher than in precipitation up to a factor of 100 [Fuzzi et al., 1996; Choularton et al., 1997; Thalmann et al., 2002; Burkard et al., 2003; Lange et al., 2003]. Acidification as a result of high SO_2 emissions was especially problematic in the border area of Germany, the Czech Republic and Poland [Lange et al., 2003; Błaś et al., 2008, 2010]. But also in North America fog was found to be a major source of acidification via hydrogen ion deposition [Schemenauer, 1986; Schemenauer et al., 1995]. The common definition of fog is a cloud in contact to ground with a meteorological visibility $\leq$ 1000 m. Therefore, cloud and fog is the same phenomenon in the context of this study.

So far, the focus of research has mainly been on areas with high atmospheric pollution, and on tropical montane cloud forests where high biodiversity is coincident with frequent occurrence of fog. However, little is known about the relevance of chemical compounds in fog in mountain areas of Switzerland, where fog is frequent during the vegetation period and where ecosystems

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tend to be adapted to low-nutrient conditions. Cloud and precipitation chemistry at higher elevations and along an altitudinal gradient in Switzerland had only been investigated in 1984/1985 at Mt. Rigi (1798 m a.s.l.) which is located 75 km NE of our main study site [Staehelin et al., 1993; Collett et al., 1993a,b].

In mountainous areas, deposition chemistry is largely influenced by topography, altitude, valley orientation and exposure to main wind directions [Rogora et al., 2006]. An assessment of atmospheric deposition in the Alpine region, mainly focusing on nitrogen deposition, can be found in Rogora et al. [2006]. They used data from previous and ongoing projects to investigate geographical variability and temporal trends of wet deposition. Although air pollution has decreased in the last decades [e.g. Barmpadimos et al., 2011], the deposition of nitrogen is still expected to be a critical factor at some sites. The role of cloud and fog deposition remains unknown, but should not be ignored, as Rogora et al. [2006] emphasize. Previous studies have shown the importance of altitudinal gradients [Miller et al., 1993], where deposition rates were found to increase with altitude, which would imply highest deposition rates at the mountain tops. The goal of our study hence was to quantify concentrations in fog water at a mountain in the Prealps (Niesen) that is prominently exposed to northerly winds from the densely populated Swiss Plateau. We hypothesize that even at such relatively remote locations substantial amounts of plant-accessible nitrogen may be deposited by fog. We expect this to be most relevant at times, when the Niesen mountain may be affected by pollutants originating within a few tens of km distance, whereas at other times its elevation is clearly above the polluted atmospheric boundary layer. To distinguish mountain top (exposed) conditions from mid-elevation conditions at the mountain, and to resolve altitudinal differences in fog chemistry, measurements were carried out in parallel at two sites at $\approx$ 2300 m and 1650 m a.s.l. (Figure 1). Moreover, additional samples were taken at the lower-elevation Lägeren Mountain site (682 m a.s.l.) to allow for a direct comparison with earlier fog water flux studies [Burkard et al., 2003]. As an outcome, the first order-of-magnitude estimate of N deposition by fog for a mountain top in the Alps was attempted in order to assess the relevance of fog as an N deposition pathway for alpine vegetation.

2. Material and Methods

2.1. Observation Sites

From 22 June to 6 October 2006 and from 23 April to 22 October 2007 (one device running until 23 November 2007) two MiniCASCC (see Section 2.2) were operated on the slope of the Niesen mountain (2362 m a.s.l.) in the Swiss Alps. The Niesen mountain is located ≈ 35 km south of Bern at the northern limit of the Central Alps (Figure 1). It is the first peak of the 22 km long northeast–southwest running Niesen ridge, and it's elevation is ≈ 1700 m above the bottoms of the surrounding valleys. The vegetation cover is subject to an altitudinal gradient and consists of mixed forests (dominated by *Fagus sylvatica* (L.) and *Picea abies* (L.) Karst.) on the southern slope up to an altitude of 1400 m a.s.l., followed by coniferous forests up to 1900 m a.s.l., and then by grassland and bare rocks.

One device (Niesen Schwandegg, S) was installed at 3 m above ground on an avalanche protection structure at 1650 m a.s.l. (46°38′32.3″ N, 7°40′03.3″ E) in a wood glade (coniferous forest) on a south-east facing slope; the second device (Niesen Kulm, K) was installed 1.5 m above ground at 2330 m a.s.l. (46°38′41.7″ N, 7°39′05.8″ E; in May 2007 moved 130 m in direction North-east to 2300 m a.s.l., 5 m above ground) on a south facing slope just below the mountain summit (alpine grassland and rocks). From 8 August 2007 to 23 November 2007 a third MiniCASCC (Niesen Kulm, B) was installed near the mountain summit (2300 m a.s.l., same location as K). These devices were supported by batteries that were exchanged during the weekly maintenance visits. Additionally, from 11 June 2007 to 23 November 2007 a wet-only precipitation sampler was installed at Niesen K.

From 7 November 2005 to 9 May 2008 one MiniCASCC was operated at the Lägeren research tower in Switzerland (47°28′42.0″ N, 8°21′51.8″ E, 682 m a.s.l.) at 35 m above ground, which is 5 m above the forest canopy (Lägeren, L). This site has already been used for fog studies by Joos and Baltensperger [1991] (winter 1986/87) and Burkard et al. [2003] (winter 2001/02), and is part of the Swiss air quality monitoring network (NABEL, since 1986). The vegetation cover is a species-rich mixed forest (dominated by *Fagus sylvatica* (L.) and *Picea abies* (L.) Karst.; see Eugster et al. [2007] for details). The tower is located at the southern slope of the Lägeren

mountain (866 m a.s.l.) which marks the northern boundary of the Swiss Plateau, ≈ 15 km northwest of Zürich (Figure 1).

From 4 April 2007 to 9 May 2008 a second MiniCASCC was installed at the same location, but at 16 m above ground level, inside the forest canopy (Lägeren, X) to assess the effect of canopy interception on fog water concentrations. At this test site, it was possible to connect both collectors to mains power using a battery charger.

2.2. Instrumentation

We used self-built MiniCASCC cloud water collectors [Michna et al., 2013] with an estimated cutoff droplet diameter of $\approx 7\ \mu$m (roughly 4 m s^{-1} measured airspeed at 2300 m a.s.l. under ambient conditions) and typical collection rates in the range of ≈ 10–20 ml h^{-1}. This collector type is a modified version of the Caltech active strand cloud water collector (CASCC, Daube et al. 1987; Demoz et al. 1996). Our collectors are battery driven, which extends the operation range to sites with no access to the power grid. With a 10×10 cm inlet they are considerably smaller than the CASCC2 [Demoz et al., 1996]. The liquid water content (LWC) was computed from collected water volume and the collection efficiency (see Michna et al. [2013] for details). The collection efficiency depends on the actual droplet spectra and is roughly 0.7 for a LWC of 100 mg m^{-3}, sampled at a nominal flow rate of ≈ 150 m^3 h^{-1}. Collection efficiency increases to 0.8 at LWC values of 300 mg m^{-3} and higher. Bulk samples of fog/cloud water were typically taken on a weekly basis.

The devices were complemented with sensors for horizontal visibility, air temperature and relative humidity, wind speed, and wind direction. Air temperature and relative humidity were measured using Rotronic Hygroclip S3 and Rotronic MP103A sensors (Rotronic AG, Switzerland), wind speed and wind direction were measured with a Young W-Monitor (R. M. Young Company, Michigan, USA). Visibility was measured with a MiniOFS Mk II sensor (Sten Löfving, Optical Sensors, Göteborg, Sweden). Data storage and collector control were done with Campbell CR510 and CR10X dataloggers (Campbell Scientific Ltd, UK). The meteorological sensors were sampled every 10 s and measurements were recorded as 10 min averages.

Precipitation was collected with the same wet-only samplers that Thalmann et al. [2002] used. The sampler contained a polyethylene bucket and was controlled by a heated water droplet sensor to open and close the lid. For minimizing heating by solar radiation, the sampling container was coated by a styrofoam layer and a reflective aluminum cover [Eugster et al., 1998; Eugster, 1999].

2.3. Sampling

Fog collection was started when visibility dropped below 300 m. In general, fog is defined by meteorological visibility ≤ 1000 m [Burkard et al., 2003]. Only dense fog, however, contains sufficient water to allow efficient collection [Eldridge, 1971]. Experience from an earlier study by Thalmann et al. [2002] showed that a threshold of 500 m to start fog collection is more adequate. Aleksic et al. [2009] and Wieprecht et al. [2005] even used much lower thresholds of 50 and 100 mg m^{-3}, respectively, in combination with an air temperature threshold of 2 and 1°C, respectively. The visibility threshold of 300 m that was finally used at our sites was chosen due to technical reasons detailed in Michna et al. [2013]. Additional thresholds were a relative humidity of > 90% and air temperature >2 °C as suggested by Baumgardner et al. [1997]. A temperature threshold was necessary since heating of the battery powered device (12 V with totally 72 Ah) was not possible. The MiniCASCCs collected fog water in 2.0 L glass bottles (see Michna et al. [2013] for more details) which were emptied every week at the Niesen sites, whereas the sampling intervals varied between 8 and 47 days at the Lägeren site but were typically within 20–35 days. Since the sample collection was not always coinciding with fog conditions, it was unavoidable to see a delay of up to several days between the last sampled fog event and sample collection. Table 1 shows the overall availability of the sampling systems. In all cases availability was better than 89%.

2.4. Analytical Procedures

After visiting the sites, the samples were filtered through a 0.45 μm nylon filter for removing particulate matter. 30 ml were filled in pre-cleaned polyethylene bottles and stored in a freezer before chemical analysis. 2 ml were stored in glass vials in a refrigerator for analysis of δ^2H and δ^{18}O values. With the remaining water, electrical conductivity and pH were measured

immediately (TetraCon WTW, Germany; respective Single Pore pH, Hamilton, Switzerland). The concentration of major inorganic anions and cations (F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-}, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Sr^{2+}) were measured by ion chromatography using a Dionex DX120 (Sunnyvale, CA, USA). In most cases the concentrations of F^- and Sr^{2+} were at or below the detection limit of our measurement device.

The water samples were analyzed for $\delta^{18}O$ and $\delta^{2}H$ values using the high-temperature carbon reduction method [Gehre et al., 2004] by coupling an elemental analyzer (TC/EA; Finnigan MAT, Bremen, D) to a Delta^{plus}XP isotope ratio mass spectrometer via a ConFlo III interface (both Finnigan MAT; Werner et al. 1999). Processing of the samples followed the procedures described by Werner and Brand [2001]; all isotope results were normalized to V-SMOW/SLAP [Coplen, 1988]. The mean uncertainty of $\delta^{18}O$ and $\delta^{2}H$ as determined from triplicates of samples were ± 0.1 and $\pm 0.7\times10^{-3}$ with 99% of all values better than ± 0.3 and ± 1.8, respectively.

2.5. Data Quality

Several blank samples collected via spraying deionized water into the fog collector were taken during and after the field operation phases to assess possible chemical contamination of the fog water collected by the sampler. Several samples were measured twice to check the precision of the analytical procedures. No signs of significant contamination of the collected water could be found. Data quality was also checked by comparing the measured and the calculated electrical conductivity of the samples. One sample had to be rejected because the measured conductivity was lower than the calculated one, whereas in all other cases the measured conductivity was equal to the calculated value, or higher. The amount of water in a few samples was too small to measure conductivity; in these cases the available water was used for chemical and stable isotope analyses only. In general, the ion balance was not even (Fig. 2), as can be expected if water soluble organic compounds are present in fog water, but are not measured (e.g., Raja et al. 2009; Collett et al. 2008; Fuzzi et al. 2002; Decesari et al. 2000; Facchini et al. 1999; Collett et al. 1999).

2.6. Isotopic Reference Data

The stable isotopes measured in fog water from our field sites are compared to a local meteoric water line (LMWL) calculated from routine measurements by the Swiss monitoring network, as

there are no direct reference values available for our sampling locations. Data from the network were obtained during the years 1995–1998 and were published in the Swiss Hydrological Atlas [Parriaux et al., 2001]. The measurement sites included in the network are Vaduz (Liechtenstein), Guttannen, Meiringen, Bern, La Brèvine, Buchs-Suhr, Grimsel, Sion, Nyon, Locarno-Monti, and Pontresina. This set of sites provides a good spatial and elevational coverage of the geographical area of interest in the vicinity of the Niesen and Lägeren mountains, but from different years.

2.7. Estimation of Occult Deposition

Direct measurements of fog water fluxes by means of eddy covariance (e.g., Vermeulen et al. 1997; Thalmann et al. 2002; Burkard et al. 2003; Klemm et al. 2005; Klemm and Wrzesinsky 2007; Westbeld et al. 2009) could not be performed at the Niesen mountain. Hence, we used the deposition model by Lovett [1984], which had been successfully applied to high-elevation forests before (e.g., by Miller et al. [1993]). There are limitations on the application of this model (see, e.g., Aleksic et al. [2009]; Klemm et al. [2005]), which means that occult deposition estimates obtained in this way should only be considered rough estimates of the order of magnitude of the true fluxes. The Lovett [1984] model is driven by: wind speed above the canopy (measured on site), fog droplet spectra (estimated from measurements done at the Lägeren site) and liquid water contents (estimated from visibility measurements), in combination with various site-specific vegetation parameters. The liquid water flux obtained from the model was then multiplied with the mean ion concentrations measured in the fog water to quantify occult deposition rates.

3. Results

3.1. Fog Characterization at the Sampling Sites

3.1.1. Fog Observations

At Niesen K, fog frequency during the sampling periods was 26.7% (673 h) in 2006 and 20.4% (1050 h) in 2007 (according to our definition using horizontal visibility below 300 m). During 43.6% and 25.9% of the hours with fog the sampling of fog water was possible, which resulted in 9 and 17 samples with sufficient amounts of fog water for analysis in 2006 and 2007, respectively. At Niesen S fog was less frequent with an average occurrence of 6.1% (161 h) in

2006 and 8.4% (366 h) in 2007. During 90.1% and 59.1% of the hours with fog sampling was possible (10 and 16 samples in 2006 and 2007, respectively). The number of days per month with fog was ≈ 20 days at the Niesen K site and ≈ 10 days at the other sites (Niesen S and the Lägeren L and X sites) (Table 2). The values observed at Niesen K compare well with sites analyzed by Wanner [1979] and with the Jungfraujoch site (3576 m), where around 17–18 days of fog were found in all months (40% of the time, Spiegel et al. [2012b]). The peak occurrence of fog was between March and June with a distinct maximum in May (≈ 22 days). Occurrence of fog, however, varies strongly among years in a way that makes comparison with studies carried out in different years problematic. As an example, Wanner [1979] found only a monthly average of 5 days with fog at the Adelboden site (1340 m a.s.l.) during the period 1932–1960, and the maximum of occurrence was in October, related to autumn thermal inversions.

At Niesen K it was only possible to capture complete fog events in a few cases during the summer months (Figure 3), since during frontal events air temperature was too low for running the collector (see also Table 2). Contrastingly, at Niesen S it was possible to sample a large number of the occurring fog events. At the Lägeren sites, sampling was always possible during summer and fall, however no radiation fog (RF)—which is the typical fog type during winter—occurred during the times of measurement. Fog preferentially occurred during the morning at all sites, and at the two Niesen sites additionally during the evening hours (Figure 4), reaching probabilities up to 40% at Niesen K site.

In mountainous areas it is often difficult to obtain representative visibility measurements, because clouds intercepting the ground can be fragmented, and periods with fog may alternate with fog-free conditions within minutes. Therefore, as a measure of the duration of fog events we only considered consecutive hours with fog (with at least 20 min presence of fog per hour). Even with this definition, fog events were relatively short. The median event duration was 2 h at the Lägeren L and Niesen S sites (average duration 3.8 h in both cases), and 3 h at the Niesen K site (average 5.6 h). The longest event duration was around 40 h at all sites. This implies that our samples are in general a collection of several fog events: at the Niesen K site 2–24 fog events contributed to each individual sample (with an average of 10.7 events). At Niesen S these numbers an average 5.5 events were covered by each sample (range 0–13 fog events, where 0 indicates that the

duration of fog was shorter than 20 min per hour). At the Lägeren L site 15.3 events were covered by each sample on average (0–53 events). However, it should be noted that the sampling interval was usually longer at the Lägeren sites than at the two other sites.

3.1.2. Estimated Liquid Water Contents

Liquid water contents (LWC) were derived from measured collection rates. Although collection rates depend on droplet size spectra, and both droplet size spectra and LWC evolve over time (e.g., Fuzzi et al. 1997; Pruppacher and Klett 2010; Price 2011), this approach should still be adequate to obtain a rough estimate of LWC. At the Niesen LWC varied from 31 to 312 mg m^{-3} (median 116 mg m^{-3} at Niesen K and 91 mg m^{-3} at Niesen S), and at the Lägeren from 63 to 206 mg m^{-3} (median 123 at 35 m a.g.l., and 110 mg m^{-3} at 16 m a.g.l.), which is consistent with previous studies at the Lägeren site [Burkard et al., 2003]. The largest variation was found at Niesen K, where both stratiform and large convective clouds with relatively high LWC are rather common. A relation between electrical conductivity (Lf) and estimated LWC could only be found at Niesen K.

3.2. Fog Chemistry Characterization

At all four sites, the most abundant inorganic ion was NH_4^+, followed by NO_3^- and SO_4^{2-} (Table 3). Altogether, these three major ions contributed between 63% (Niesen K) and 78% (Lägeren, L) to the average ion balance, with a maximum of 94%. In general, these three dominant ions were present in remarkably lower concentrations at the two Niesen sites, compared to the two Lägeren sites. The differences among sites were significant for the Niesen K site in comparison with both Lägeren sites, and between Niesen S and Lägeren X ($p < 0.05$, Wilcoxon rank sum test). Furthermore, Ca^+, Na^+, and PO_4^{3-} (the latter at the Lägeren X site) contributed substantial amounts to the ion balance. No relationship between electric conductivity (as measure for the total ion loading) and the contribution of these major three ions could be found.

Ion concentrations varied considerably between sampling periods, sites, and also between the two sampling seasons. In general, ion loadings were lower in 2007 than in 2006 at all sites with two seasons of available data. Electric conductivity showed high variability, but only the differences between the two Lägeren sites and both Niesen sites were significant (Table 4).

All major ions (Table 3) showed concentrations similar to what Burkard et al. [2003] reported from fog associated with atmospheric instabilities (FAI) but were substantially lower than what they reported from radiation fog (RF) events. This difference can be attributed to the inter-annual variability and the fact that our equipment was unable to capture RF, which often was associated with temperatures below the operation limit of our fog collectors. This interpretation is supported by the average wind direction during collection times at the Lägeren which was SW in nearly all cases, whereas RF events are in most cases associated with easterly winds.

Non-sea salt contributions of SO_4^{2-} and Ca^{2+} as indicators for pollution were calculated using the same method used by, e.g., Lu et al. [2010] or Watanabe et al. [2006], with Na as tracer for marine source [Morales et al., 1998]:

$$[\text{nss-}SO_4^{2-}] = [SO_4^{2-}] - 0.2455\,[Na^+]\,, \tag{1}$$

$$[\text{nss-}Ca^{2+}] = [Ca^{2+}] - 0.0373\,[Na^+]\,. \tag{2}$$

Since Na^+ can also originate from soil dust and biomass combustion [Lu et al., 2010; Millet et al., 1996], the non-sea salt contributions might be underestimated. A comparison between our sampling sites showed no statistically significant differences in the Na^+ concentrations ($p < 0.05$, Wilcoxon rank sum test), therefore such additional sources are not likely to contribute large amounts to the ion budget, and hence were not treated explicitly in our calculations.

The relationship between nss-SO_4^{2-} and Lf, respective nss-Ca^{2+}, is shown in Figure 5. In the case of nss-SO_4^{2-}, the relative contributions varied between 0.7 and 1.0. Lowest values were found at Niesen K, followed by Niesen S, while at both Lägeren sites the contribution was normally between 0.95 and 1.0. At the Niesen sites, a positive correlation between nss-SO_4^{2-} and Lf was found, which was, however, only significant ($p < 0.01$, Wilcoxon rank sum test) at Niesen K. For nss-Ca^{2+} the same relationships as for nss-SO_4^{2-} were found, and in addition the correlation between nss-Ca^{2+} and Lf was significant at Niesen S.

The very high Ca^{2+} concentrations, primarily observed at low LWC in the first samples collected at the Niesen K site in 2007, can be explained by the very unusual warm and dry April 2007 (only 33% of the long-term precipitation amount was recorded at the MeteoSwiss site Adelboden, and

temperature exceeded the long-term average of the period by 5.9°C; MeteoSchweiz [2007]) which led to unusually large amounts of mineral dust in the air. Since the bedrock of the Niesen mountain range consists of limestone and slate, the high Ca^{2+} concentration levels and consequently also pH [Fisak et al., 2002; Lakhani et al., 2007] in fog water may stem from dust particles of local origin, a phenomenon that was also observed by Cini et al. [2002] at the Vallombrosa mountain site in the Tuscal Apennines.

The large variation of nss-SO_4^{2-} at the two Niesen sites indicate both, long-range and local influence. Given their altitude which is typically in the upper part of the atmospheric boundary layer, the Niesen sites also observed events with characteristic oceanic (long-range) influence. Contrastingly, at the Lägeren sites >90% of the higher SO_4^{2-} concentrations was nss-SO_4^{2-}, which means that the source influencing the fog at the Lägeren site must be from local or regional sources, rather than long-range transport.

3.2.1. Fog Chemistry Differences Between Sites

The relative composition of the fog water was similar at all sampling sites (Table 4). Concentrations of the major inorganic ions were however generally lower at Niesen K than at Niesen S, and lower at Lägeren L than at Lägeren X. The concentrations of the major ions NH_4^+, NO_3^-, and SO_4^{2-} at Niesen K were ≈ 30–55% lower concentrated than at Niesen S. However, these differences were not statistically significant. Ca^{2+} concentrations were significantly higher at Niesen K. Niesen K concentrations were significantly lower than at Lägeren L ($p < 0.05$) with only 10–20% of those found at Lägeren L. The difference between Niesen S and Lägeren L was somewhat less pronounced with 20–60% of Lägeren L concentrations found at Niesen S, but still statistically significant (Wilcoxon rank sum test). Concentration ratios between the two Lägeren sites L (upper) and X (lower) ranged between 0.5 (NH_4^+) and 1.3 (NO_3^-), but were not statistically significant.

Na^+ concentrations did not show any dependency on sampling location. All weighted means were around 40 μeq l^{-1}, except for the Lägeren L site, where the mean was nearly 80% higher. At both Niesen sites a seasonal pattern was found in 2007, but not in 2006, with lower concentrations during the summer months and higher concentrations in spring and autumn.

3.3. *Rainwater Chemistry Compared to Fog Water*

A precipitation total of 619.5 mm was collected at Niesen K (3.75 mm d^{-1}), which is 75% of the sum collected at the nearest MeteoSwiss SwissMetNet site Adelboden during the same period (18 km southwest, 1340 m a.s.l.). The probable under-sampling of precipitation at the Niesen K site is probably due to the different sampling techniques. At Niesen K a wet-only sampler was used, which probably under-samples snow when snowfall coincides with stronger winds. Moreover, the heater of the precipitation sensor may not perform perfectly under such conditions, and if snow on the sensor is not melted immediately, then the lid of the wet-only sampler may not have opened instantaneously with onset of frozen precipitation at Niesen K.

Electrical conductivity as a measure of the total ion loadings was found to be low with values varying from 4.3 to 21.3 μS cm^{-1}, while the range of fog water at Niesen K during the same period was 14.1–49.9 μS cm^{-1} (Table 5). In weighted means, the corresponding values amount to 32.5 μS cm^{-1} in fog water and 8.5 μS cm^{-1} in rainwater, which translates to a ratio of 3.5. At the lower elevated site Niesen S electrical conductivity varied from 23.1 to 206 μS cm^{-1} with a weighted mean of 29.9 μS cm^{-1} which is 7.1 times that of rainwater at Niesen K ($\approx$ 1 km W). pH varied from 5.1 to 7.7 in rainwater (weighted mean 6.5) which is slightly higher than the expected natural CO_2 equilibrium background value [Seinfeld and Pandis, 1998], while the corresponding values for the two fog collecting sites were in the range of 5.8–7.3.

The relative composition of fog- and rainwater was similar, the major ions in fog water were NH_4^+, NO_3^-, SO_4^{2-}, and constitute 72.2 and 75.1% of the total ion loadings for Niesen K and Niesen S, respectively. In rainwater, these ions contribute only to 32.2%, abundant additionally contributing ions were Na^+ (38.4%), and Ca+ (11.7%). However, in absolute numbers, the concentrations of Na^+ are very similar with 44.6 μeq l^{-1} in rainwater and 42.5, respectively 41.5 μeq l^{-1} in fog water. For the other major ions, absolute concentrations in terms of weighted means are considerably higher. Significant differences to the Niesen K site were found for all ions except Na^+.

3.4. *Stable Isotopologue Ratios*

δ^2H and δ^{18}O varied from –65.6 to –25.3×10^{-3}, respectively from –10.1 to –5.0×10^{-3} at the Niesen K site. At the Niesen S site, the variations were in the ranges –65.4 to –27.7×10^{-3} (δ^2H) and –1.7 to –4.9×10^{-3} (δ^{18}O) (Figure 6). At the Lägeren sites δ^2H varied from –72.8 to –29.4×10^{-3} and δ^{18}O –9.1 to –5.5×10^{-3} (not shown), which is similar to both Niesen sites, but with a tendency towards slightly more negative values. Our δ-values are in the range of a previous study in Central Europe [Spiegel et al., 2012a], but substantially more negative than in the tropics (e.g., Scholl et al. 2011).

A comparison between Niesen K and Niesen S generally shows more negative values in δ^2H and δ^{18}O at the higher elevated site (Figure 6). This might reflect the fact, that at the Niesen S site—apart from thermal inversions during autumn—stratus fractus clouds are often observed, which develop from re-condensation during frontal passages.

Cloud water lines (CWL) were calculated by orthogonal regression of δ^2H vs. δ^{18}O (Figure 6). At the Niesen sites the best fit was $\delta^2\mathrm{H} = 8.77\,\delta^{18}\mathrm{O} + 18.30 \times 10^{-3}$ (n=17) at Niesen K, and $\delta^2\mathrm{H} = 10.01\,\delta^{18}\mathrm{O} + 24.33 \times 10^{-3}$ (n=15) at Niesen S, which is above the global meteoric water line (GMWL, Craig 1961) but in the range of the local meteoric water line LMWL ($7.19\,\delta^{18}\mathrm{O} - 2.0 \times 10^{-3}$, Parriaux et al. 2001), although slightly steeper. The CWL at Niesen K appears to be closer to the LMWL than the CWL of the Niesen S site. At the Lägeren sites the CWLs are almost identical (at the upper site $\delta^2\mathrm{H} = 13.18\,\delta^{18}\mathrm{O} + 48.63 \times 10^{-3}$, n=8; at the lower site $\delta^2\mathrm{H} = 13.85\,\delta^{18}\mathrm{O} + 50.85 \times 10^{-3}$, n=8). They are much steeper than the LMWL, but associated with a large uncertainty because they are only based on a very limited number of observations.

3.5. *Estimation of Occult Deposition*

Since the Niesen K site is influenced by a strong vertical wind component as was visually observed during convective situations, and moreover the topography at this site is complex, we only present ranges of occult deposition fluxes in the following.

Both, occult deposition of sulphur and nitrogen as estimated by the Lovett [1984] deposition model for the Niesen K site, lie in the range between 1.0 and 15 mg $\mathrm{m}^{-2}\,\mathrm{s}^{-1}$ during periods with

fog. Annual inputs hence strongly depend on fog frequencies. The annual mean fog frequencies at the three sites Niesen K, Niesen S, and Lägeren L, as calculated from the two field periods, are 25.7%, 7.3%, and 6.3%. The corresponding fog water flux range from 8 to 121 mm at Niesen K, 2–35 mm at Niesen S, and 2–30 mm at Lägeren L. At the Lägeren site directly measured liquid water fluxes are available from 323 days in 2001/2002 [Burkard et al., 2003]. They measured fog water fluxes of 2.7 mm by FAI and 7.3 mm by RF, which is in a comparable range as was estimated in this study.

At the Niesen K site, the estimated deposition rates for NH_4^+, NO_3^-, and SO_4^{2-} amount to 1.2–17.5, 0.7–10.5, and 0.6–8.7 meq m^{-2} yr^{-1}, respectively. Referenced to precipitation, occult deposition rates are estimated to add an additional +80%, +70%, and +48%, respectively, to the three major ion fluxes. At the Niesen S site, occult deposition rates amount to 0.5–8.8, 0.4–7.0, and 0.2–3.7 meq m^{-2} yr^{-1} for NH_4^+, NO_3^-, and SO_4^{2-}. With respect to precipitation inputs (measured at Niesen K) this is an addition of +40%, +47%, and +20% by occult deposition. At the Lägeren site, the estimated deposition rates are 1.7–25.4, 0.7–10.4, and 0.7–10.0 meq m^{-2} yr^{-1}.

4. Discussion

4.1. Fog Characterization at the Sampling Sites

At the two Niesen sites, fog is typically associated either with frontal systems or convective cloud formation during summer (FAI—fog associated with atmospheric instabilities, see Burkard et al. [2003]). Radiation fog (RF) may occur at the Niesen S site (1680 m a.s.l.), however such events are rare. From Bachmann and Bendix [1993] and Wanner [1979], it can be estimated that in less than 5% of the inversion cases which lead to RF over the Swiss Plateau the altitude of the stratus top is above than the Niesen S site. Therefore, at the Niesen K site (2300 m a.s.l.) the possibility of occurrence of radiation fog can almost be excluded. During convective situations, the Niesen K site may be above the cloud base and therefore within cumulus and cumulonimbus clouds, whereas cloud occurrence at the Niesen S site is likely to be caused by stratus fractus clouds during these cases.

With its altitude of 682 m, the Lägeren research site, located roughly 250 m above the valley floor, remains within the lower part of the atmospheric boundary layer prone to RF. During temperature

411 inversions in wintertime, which occur when an anticyclone is located east of Switzerland, the
412 Lägeren site is often located within a stratus deck which covers the lower elevations of the Swiss
413 Plateau [Wanner, 1979]. In the warm season, fog is therefore mostly associated with stratus and
414 stratocumulus clouds (FAI). Radiation fog was only likely to occur at the Lägeren sites during the
415 start and end of the sampling periods, and at the Niesen S site at the end of the sampling period.

416 *4.2. Fog Chemistry Characterization*

417 A comparison between our measurements and previous studies at different locations is shown in
418 Table 6. Compared to previous studies at the Lägeren site [Burkard et al., 2003; Joos and
419 Baltensperger, 1991] we found substantially lower concentrations of the major ions NH_4^+, NO_3^-,
420 SO_4^{2-} during both radiation fog (RF) and fog associated with atmospheric instabilities (FAI)
421 distinguished by Burkard et al. [2003]. Conductivity at Niesen S was 10% of that reported for RF
422 at the Lägeren research site (12% for Niesen K). In FAI events, conductivity was 25% at Niesen S
423 (30% at Niesen K).
424 Lf at Lägeren L was 28% higher than what Burkard et al. [2003] had found. This can be
425 explained by higher concentrations of NH_4^+ (+39%), since NO_3^- (–44%) and SO_4^{2-} (–1%) show
426 lower concentrations.
427 In 1990/1991 a comparable study was conducted at Mt. Rigi, which is approximately 75 km NE
428 of the Niesen [Staehelin et al., 1993; Collett et al., 1993b]. In contrast to the present work, they
429 investigated winter and spring fog events at elevations of 1030 and 1620 m a.s.l. [Staehelin et al.,
430 1993]. At similar altitude, we found 61% lower concentrations of NH_4^+, and 76% respectively
431 77% lower concentrations of SO_4^{2-} and NO_3^- at the Niesen S site, whereas LWC at both Niesen
432 sites was in the same range as Collett et al. [1993b] found at Mt. Rigi.
433 Inter-annual variability of the major ions at Niesen was on the order of a factor of two, which
434 compares well with the variability found at Whiteface Mountain (NY; Aleksic et al. [2009]). This
435 variability can be explained by two main factors, namely a year-to-year variation in
436 meteorological conditions [Aleksic et al., 2009], and a large variability in ion concentrations at
437 the cloud base.

4.3. Rainwater Chemistry Compared to Fog Water

The relative ionic composition of precipitation collected at Niesen K was very similar to that measured at the Lägeren site by Burkard et al. [2003] in 2001/2002. But absolute concentrations at Niesen K were generally only half those at Lägeren, except for Na^+ which showed concentrations at Niesen K that were 1.5× those at Lägeren. This indicates a somewhat larger marine influence at the Niesen compared to Lägeren, which can be explained by the difference in location and altitude, with the Niesen mountain being located 100 km SW of Lägeren and 1500 m higher in elevation.

The highest ionic enrichment factors in fog compared to rain were found for PO_4^{3-} (11.1) and NH_4^+ (8.8), followed by NO_3^-, NO_2^-, and SO_4^{2-} (Table 4). This compares well with Burkard et al. [2003] and Thalmann et al. [2002], and indicates that the local influence of ammonia emissions from livestock farming, the dominant farming type in the Niesen region, and regional NO_2 emissions does not differ dramatically at the Niesen site compared to the Lägeren site.

The relative depletion of Na^+ and Cl^- in fog water compared to precipitation is a sign of large-scale influence from the North Atlantic [Burkard et al., 2003], since there are no relevant sources of Na^+ other than the ocean upwind of mount Niesen. In seawater, the Cl^-/Na^+ ratio is roughly 1.17 [Millet et al., 1996]. At our sites, the Cl^-/Na^+ was well below seawater values, decreasing from 0.87 at Lägeren X to 0.26 at Niesen K. Most likely, Cl^- was substituted by NO_3^- and SO_4^{2-} in the ionic balance [Millet et al., 1996].

4.3.1. Estimation of Wet Deposition

Wet deposition rates at Niesen K were 22, 15, and 18 meq m^{-2} yr^{-1} for NH_4^+, NO_3^-, and SO_4^{2-}, respectively, if the values from Table 5 are scaled up to a full-year period. This is roughly 20–50% less than what Rogora et al. [2006] reported from years 2000–2001 at a site 11 km NE (Beatenberg, 1511 m a.s.l.) of Niesen with calculated wet deposition rates of 29, 27, and 22 meq m^{-2} yr^{-1}. Ion concentrations in rain at the Niesen are also quite similar to those found at Lägeren in an earlier study by Burkard et al. [2003], except for higher Na^+ values. As in fog Na^+ in rain is a tracer for sea spray influence and may be an indication that rainfall at higher elevation (Niesen) is more strongly influenced by orographic precipitation from clouds advected from the

Atlantic than by precipitation at Lägeren, where convective precipitation is more important, with a higher share of water recycled from land surface evaporation and hence lower Na^+ concentrations.

4.4. Stable Isotopologue Ratios

Solutes in fog water are rather of local and short-distance origin and not the result of transport by advected clouds over longer distances. The slopes of the isotopic cloud water mixing lines (CWL) tend to be steeper than the general GMWL. The steepness of the line is a function of temperature with a steeper slope at lower temperature [Clark and Fritz, 1997], which is partially consistent with the fact that mountain regions see colder clouds than lower elevations and lower latitudes. In equilibrium processes the slope is typically around 8.0 [Dansgaard, 1964], and lower values result from evaporation processes, whereas condensation leads to steeper slopes. At both Niesen sites the slopes were clearly > 8, indicating the relevance of condensation processes, which is an agreement with the role that convective formation of clouds play. The slopes of individual events as derived from paired samples from Niesen K and S (Figure 6) show that spring and summer events are more likely affected by condensation processes (slope > 8), whereas events in autumn are almost parallel to the GMWL and hence rather reflect equilibrium processes in the fog.
The distance from the GMWL is interpreted as an indicator for the relative humidity at the place where ocean water evaporated and is expressed as the deuterium excess $d = \delta^2\mathrm{H} - 8 \cdot \delta^{18}\mathrm{O}$, with $d = 10$ for the GMWL [Dansgaard, 1964]. At mount Niesen, d ranged from 0 (one spring sample from Niesen K) to 18 (autumn samples; Figure 6). This means that in spring fog originated from equilibrium evaporation over the ocean (at 100% relative humidity), and it is not surprising that the highest values in autumn in indicate less damp conditions where evaporation of ocean water occurred at around 70% relative humidity (see Clark and Fritz 1997).

4.5. Estimation of Occult Deposition

Although we found very low concentrations of NH_4^+, NO_3^-, and SO_4^{2-} in fog water compared to other studies (Table 6), estimated deposition rates at the Niesen K site are likely to reach values as found at lower elevated sites in polluted areas, such as the Lägeren site. The two Niesen sites

show an altitudinal gradient with increasing deposition rates with increasing elevation, which can be explained by a 3.5 times higher fog occurrence while the concentrations at are 1.5–2.3 times those of the lower elevated site. This result is consistent with previous studies (e.g., Miller et al. 1993; Kalina et al. 2002). Since occult deposition by fog was not measured directly, the ion loads had to be multiplied with estimated fog water fluxes from comparable sites.

Deposition rates on the order of ≈ 10 and ≈ 15 mg m^{-2} s^{-1} were found by Klemm et al. [2005] and Klemm and Wrzesinsky [2007] for the typical event of a 10 month fog observation period at the Waldstein site in Germany. A somewhat lower rate of 4.9 mg m^{-2} s^{-1} was measured by Thalmann et al. [2002] at the same site. Since the top of the Niesen mountain is above the local tree-line, the even lower fog water flux rate of 1.7 mg m^{-2} s^{-1} measured over agricultural vegetation on the Swiss Plateau [Thalmann et al., 2002] may be the most realistic basis to estimate occult deposition at Niesen K, whereas at Niesen S—situated below the tree-line—the actual occult deposition might actually be higher than what was estimated here.

4.6. Total Nitrogen Deposition

Occult nitrogen deposition by fog water was estimated at 0.3–3.9 kg N ha^{-1} yr^{-1} at Niesen K, and at 0.1–2.2 kg N ha^{-1} yr^{-1} for Niesen S. Precipitation contributes ≈ 5.2 kg N ha^{-1} yr^{-1}. Dry nitrogen deposition as taken from Eugster [1999] at a representative site 50 km NW of the Niesen (same approach as Burkard et al. 2003) amounts to roughly 15 kg N ha^{-1} yr^{-1}. All components together yield an estimated 20–24 kg N ha^{-1} yr^{-1}, which is clearly above the expected critical load of 15 kg N ha^{-1} yr^{-1} [Rihm, 1996]. Although gaseous dry deposition already may be sufficient to reach or exceed the critical load, fog water likely is a relevant source for additional eutrophication at the Niesen, adding up to 16% to the total nitrogen deposition in our rough estimate. If measures can be taken to substantially reduce gaseous dry deposition in the future, then it would be meaningful to obtain a better quantitative estimate of occult deposition by fog via direct eddy covariance measurements.

5. Conclusions

Extended summertime fog events were investigated at two different mountains at 4 measurement sites. The main fog types observed were fog associated with atmospheric instability and convective clouds. Dominant inorganic ions in fog water were NH_4^+, NO_3^-, and SO_4^{2-} at all sites. Referenced to precipitation, the enrichment factors in fog water were in the range 5 to 9 at the highest site Niesen K. The ion loadings in fog water during summertime at the two Alpine sites were considerably lower than at lower elevations on the Swiss Plateau. Lowest ion concentrations were found at Niesen K (2300 m a.s.l.). Highest concentrations, observed at the Lägeren sites, were up to 7 times larger. The inter-annual variability of both, ion concentrations and fog occurrence, was high, namely because of the high variability in seasonal weather patterns. From fog frequency and typical fog water flux rates, we estimated occult nitrogen deposition by fog water. This pathway might contribute up to 16% to total N deposition, which confirmed our hypothesis that even at such relatively remote locations a substantial amount of plant-accessible nitrogen is deposited by fog. The vast share of N deposition (63%), however, most likely stems from gaseous (and to a much lower extent particulate aerosol) dry deposition during the much more frequent fog-less and rainless periods, similar to what would be expected at lower elevations in Switzerland [Eugster et al., 1998]. This similarity is a result of two elevational gradients with different signs: ion concentrations in fog water clearly decrease with altitude, whereas fog frequency and liquid water inputs increase with altitude. In combination, our order-of-magnitude estimate of occult deposition by fog is expected to be a non-negligible pathway of N input also for alpine vegetation with plant species that are vulnerable to elevated N deposition rates.

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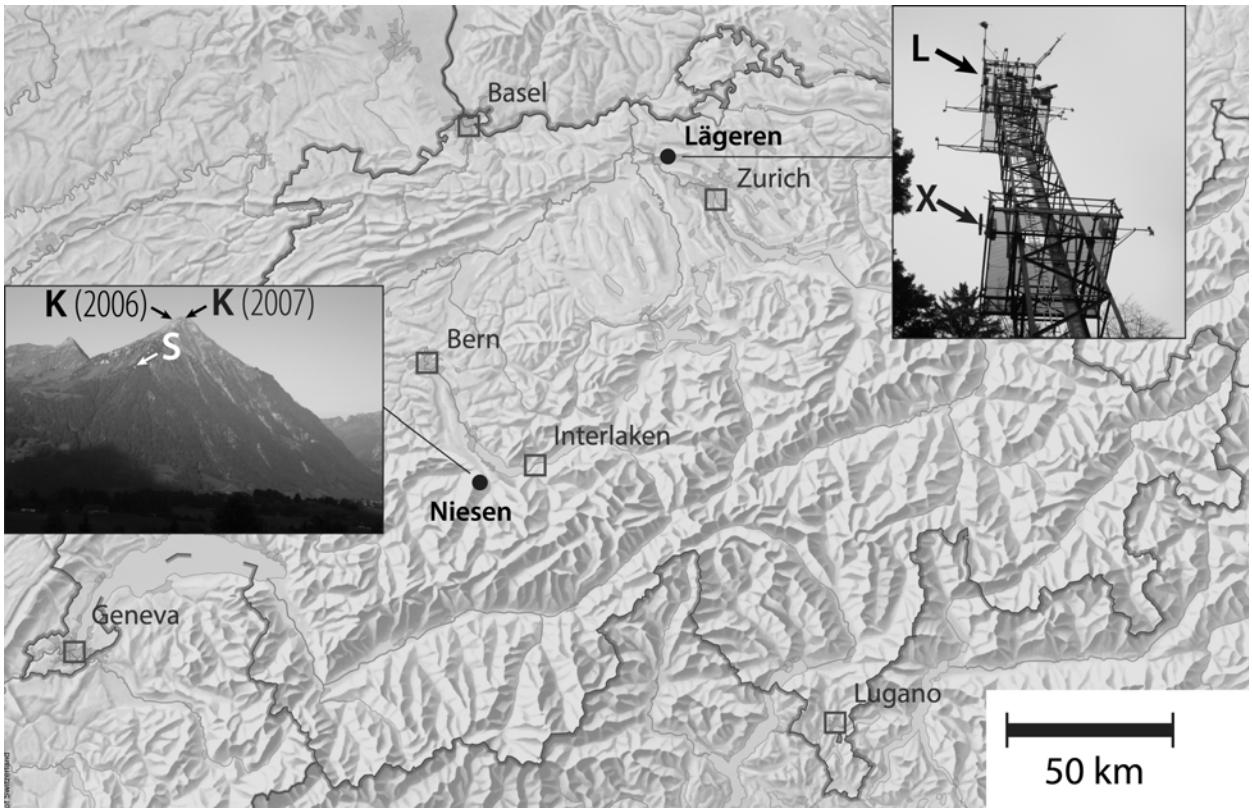

Figure 1: Localities and sites. Base map © swisstopo.

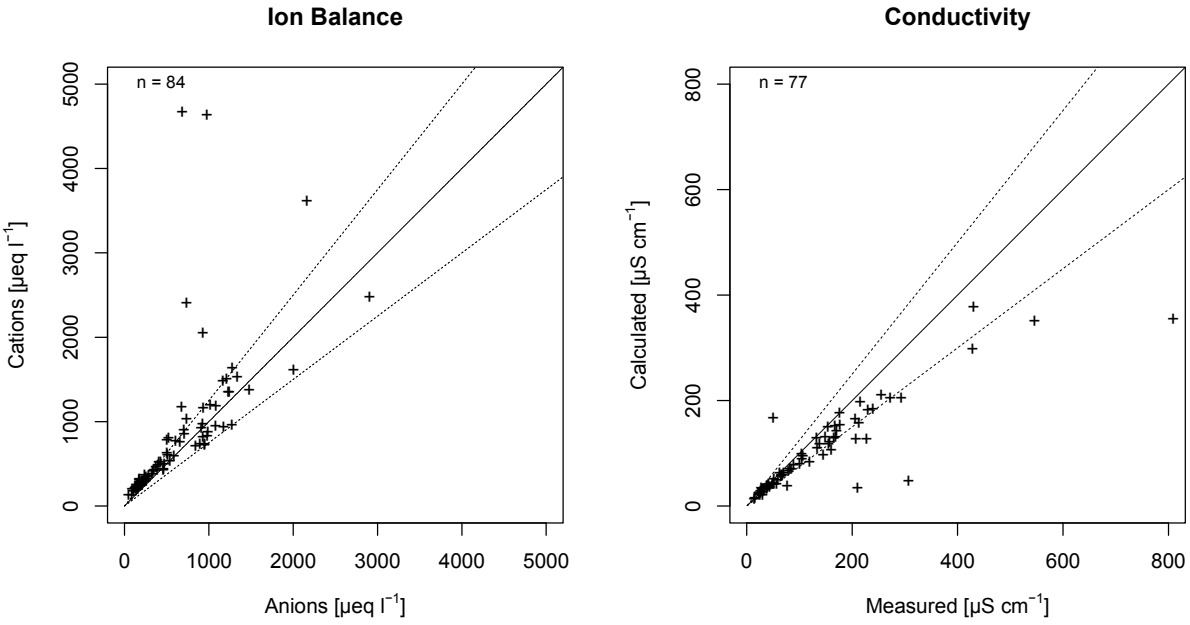

Figure 2: Fog water ion balance (left) and electrical conductivity (right) for all sites for the seasons 2006 and 2007.

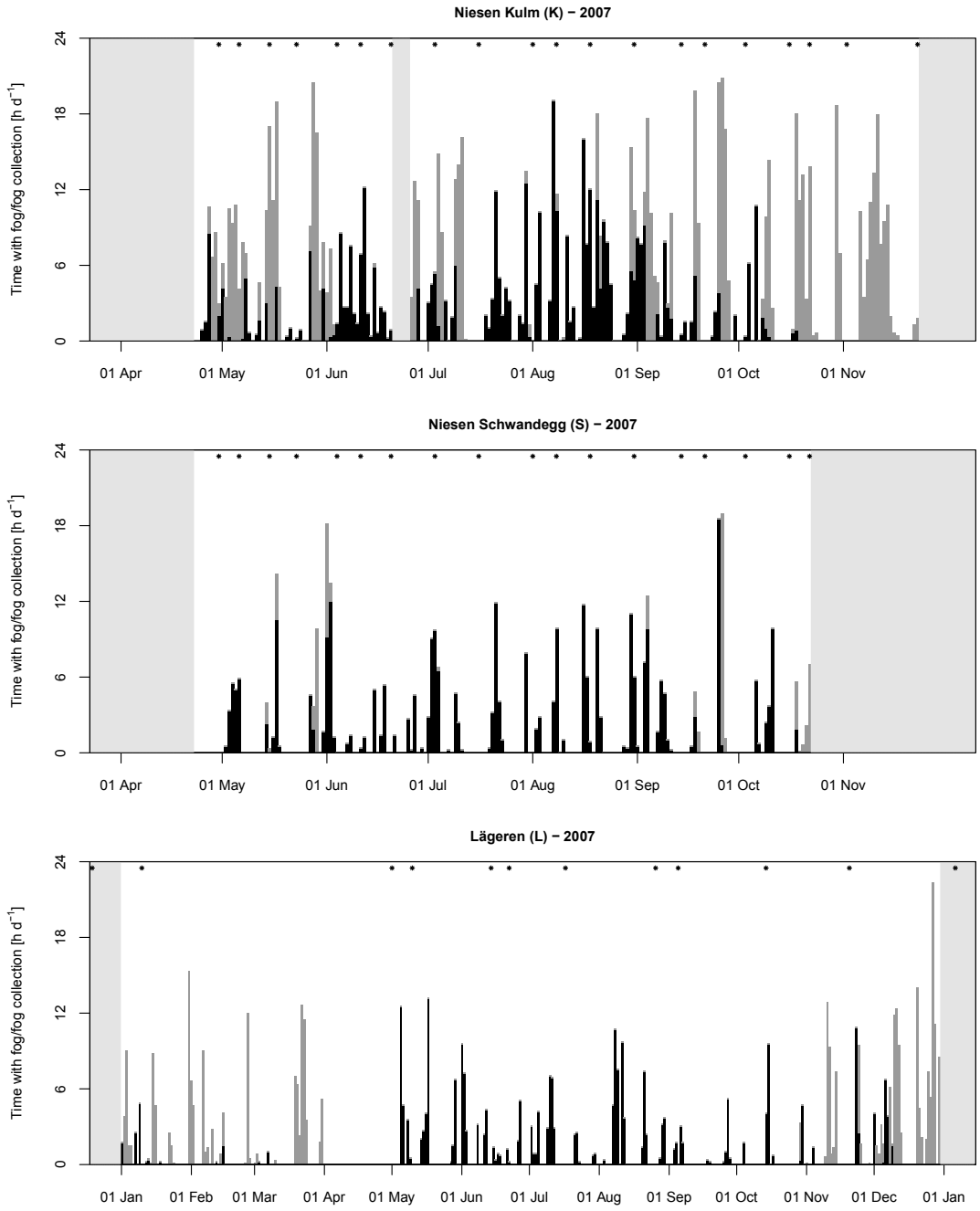

Figure 3: Hours per day with fog (grey bars) and fog collection (black bars) at the Niesen Kulm (K), Niesen Schwandegg (S), and Lägeren (L) site for the sampling period in 2007. The shaded areas denote missing meteorological data, and asterisks indicate when samples were taken, which occasionally took place during a period where the fog collector was active, but meteorological data were not correctly recorded.

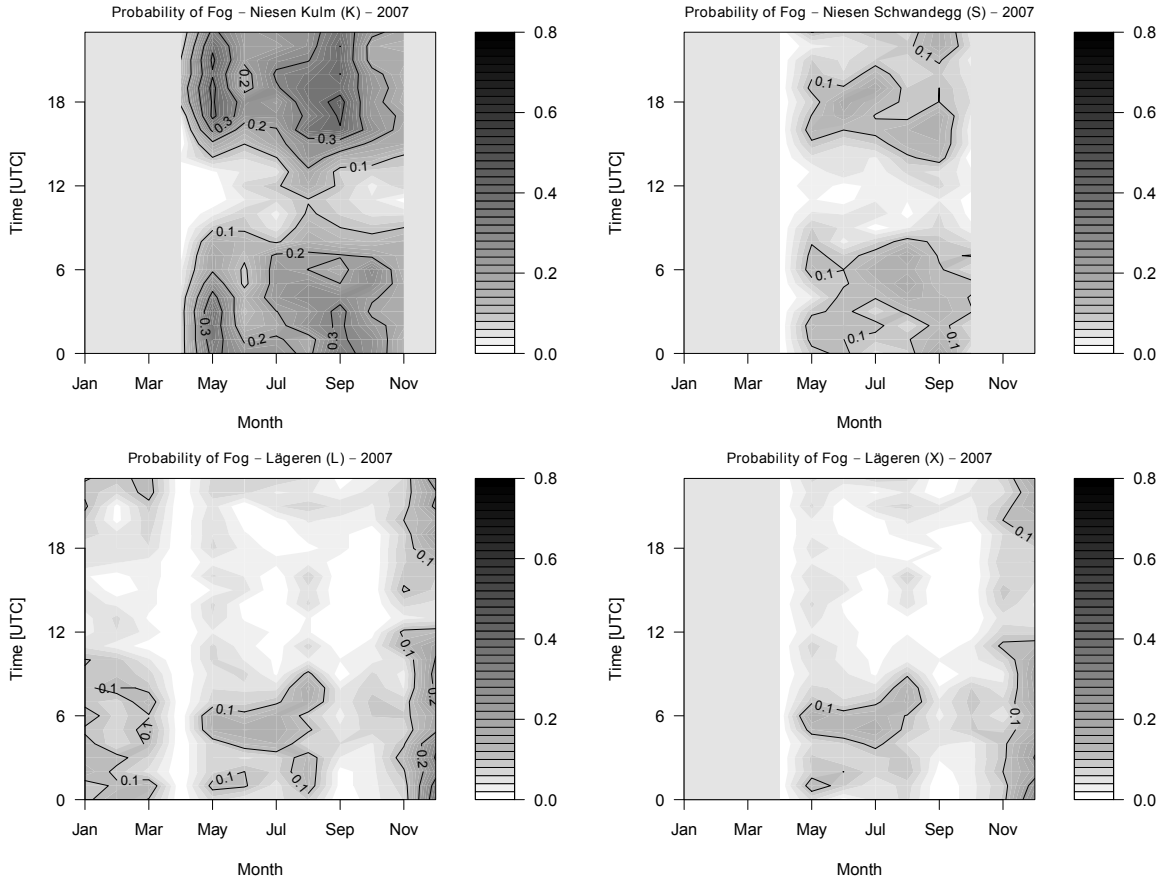

Figure 4: Probability of occurrence of fog as a function of hour of day and month of year 2007 for the sites Niesen Kulm (K), Niesen Schwandegg (S), Lägeren (L, 35 m height; and X, 16 m height). Isolines are plotted at probability intervals of 0.1 (i.e., 10% increments). Shaded areas in light grey denote not available data.

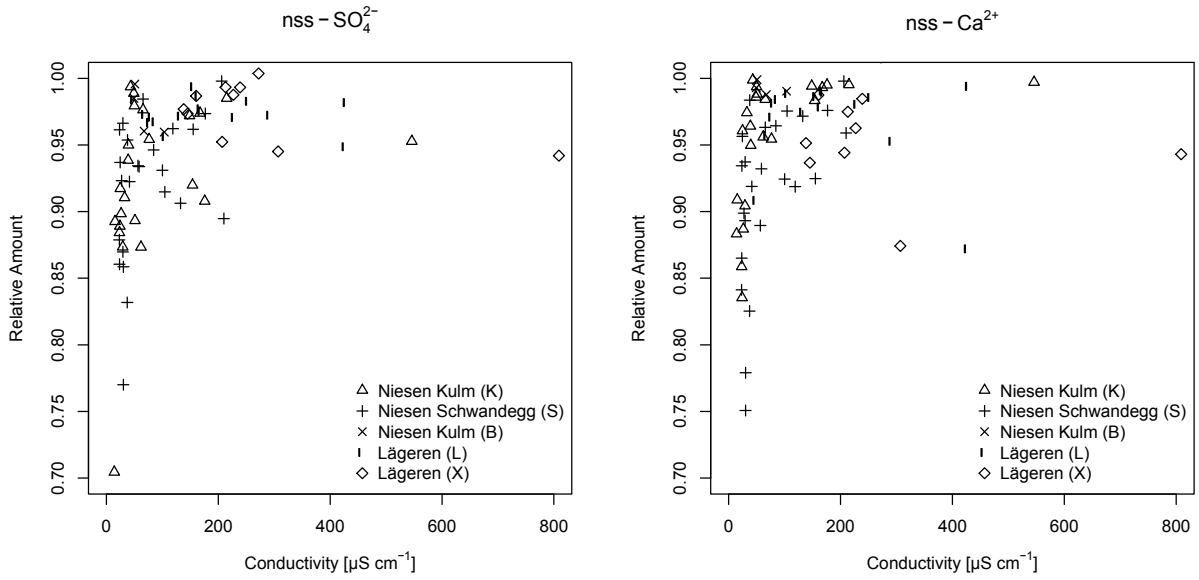

Figure 5: Relationships between electrical conductivity (Lf) and the relative amounts [nss-SO_4^{2-}]/[SO_4^{2-}] (left), and [nss-Ca^{2+}]/[Ca^{2+}] (right), respectively (measurements from 2006 and 2007).

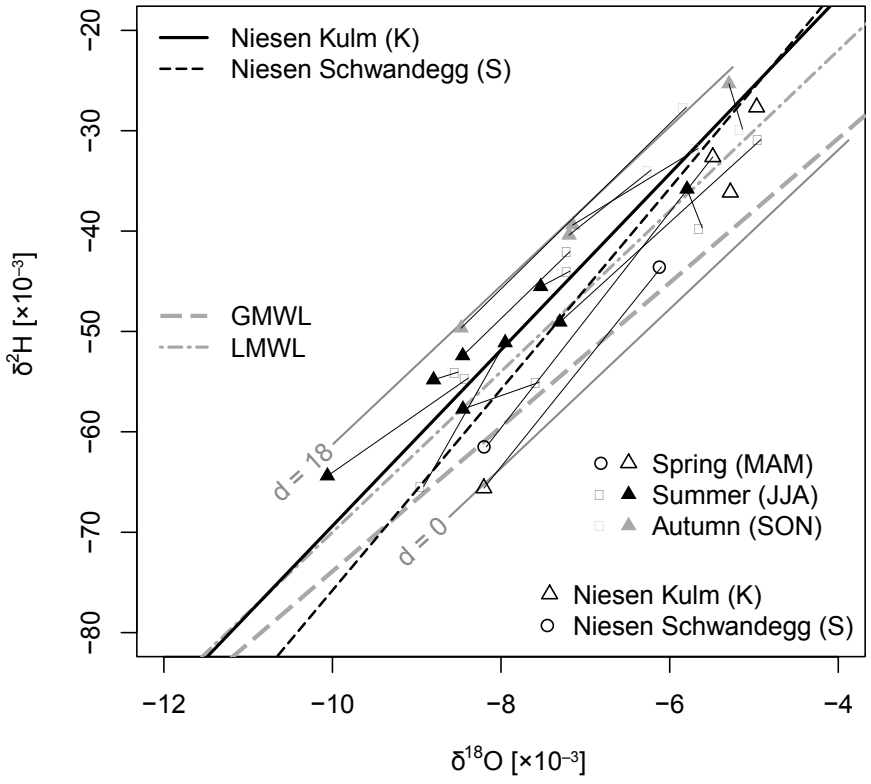

Figure 6: Isotopic ratios of δ^2H vs. δ^{18}O in fog water at the two Niesen sites, and cloud water lines (CWL) obtained via orthogonal regression fits. For reference, the global meteoric water line (GMWL) and the local meteoric water line (LMWL) for the Swiss monitoring network are also shown. Pairs of samples (same time period at different locality) are connected with a thin black line. The two lines for deuterium excess of d = 0 and 18 $\times 10^{-3}$ are shown with gray lines. The climatological seasons conform to the meteorological spring (March, April, May), summer (June, July, August), and autumn (September, October, November). For details see text.

Table 1: Site runtime and data availability for the five MiniCASCC (K, B, S, L, X) and for the wet only sampler (R).

Site	Runtime [days]			Availability [%]		
	2006	2007	2008	2006	2007	2008
Niesen Kulm (K)	105	214	–	89.4	96.3	–
Niesen Kulm (B)	–	84	–	–	94.2	–
Niesen Schwandegg (S)	110	182	–	96.3	99.6	–
Lägeren (L)	272	365	233	99.9	99.9	99.9
Lägeren (X)	–	265	233	–	99.2	99.5
Niesen (R)	–	165	–	–	90.1	–

Table 2: Fog statistics at the sites Niesen Kulm (K), Niesen Schwandegg (S), Lägeren (L), and Lägeren (X) for the extended summer season 2007. Percentages of the relative fog occurrence are given with respect to the full length of each month, whereas percentages of sampled fog relate to the percentage of the time with fog (i.e. visibility < 1000 m) that the fog sampler was running (based on the threshold value given in the text).

Niesen Kulm (K)									
	Unit	*Apr*[a]	May	Jun	Jul	Aug	Sep	Oct	*Nov*[a]
Hours with fog	h	*31.3*	187.3	100.3	135.3	176.5	187.2	134.7	*97.0*
Relative fog occurrence	%	*4.4*	25.2	13.9	18.2	23.7	26.0	18.1	*13.5*
Sampled fog	%	*41.0*	17.9	65.0	54.4	84.1	30.5	16.2	*0.0*
Hours per day with fog	h day^{-1}	*1.0*	6.0	3.3	4.4	5.7	6.2	4.3	*3.2*
Days with fog occurrence	days	*6.0*	22.0	21.0	22.0	20.0	20.0	15.0	*13.0*
Niesen Schwandegg (S)									
	Unit	*Apr*[b]	May	Jun	Jul	Aug	Sep	*Oct*[b]	Nov
Hours with fog	h	*0.0*	60.0	57.0	63.8	68.5	79.0	*37.8*	–
Relative fog occurrence	%	*0.0*	8.1	7.9	8.6	9.2	11.0	*5.9*	–
Sampled fog	%	–	71.1	81.6	99.5	100.0	67.3	*63.4*	–
Hours per day with fog	h day^{-1}	*0.0*	1.9	1.9	2.1	2.2	2.6	*1.2*	–
Days with fog occurrence	days	*0.0*	11.0	12.0	11.0	12.0	11.0	*9.0*	–
Lägeren (L)									
	Unit	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
Hours with fog	h	0.0	51.3	40.5	34.8	54.8	14.8	23.8	56.0
Relative fog occurrence	%	0.0	6.9	5.6	4.7	7.4	2.1	3.2	7.8
Sampled fog	%	–	100.0	100.0	100.0	100.0	100.0	87.4	26.2
Hours per day with fog	h day^{-1}	0.0	1.7	1.4	1.1	1.8	0.5	0.8	1.9
Days with fog occurrence	days	0.0	9.0	12.0	12.0	10.0	6.0	6.0	10.0
Lägeren (X)									
	Unit	*Apr*[c]	May	Jun	Jul	Aug	Sep	Oct	Nov
Hours with fog	h	*0.0*	50.0	34.0	32.0	46.3	11.2	22.3	52.5
Relative fog occurrence	%	*0.0*	6.7	4.7	4.3	6.2	1.6	3.0	7.3
Sampled fog	%	–	100.0	100.0	100.0	100.0	100.0	100.0	44.4
Hours per day with fog	h day^{-1}	*0.0*	1.6	1.1	1.0	1.5	0.4	0.7	1.8
Days with fog occurrence	days	*0.0*	10.0	9.0	12.0	10.0	5.0	5.0	7.0

[a] Data available from 23 Apr 2007 to 23 Nov 2007
[b] Data available from 23 Apr 2007 to 22 Oct 2007
[c] Data available from 11 Apr 2007

Table 3: Ion concentrations [μeq l^{-1}], pH and conductivity [μS cm^{-1}] at the sites Niesen Kulm (K), Niesen Schwandegg (S), Lägeren (L) (seasons 2006 and 2007), and Lägeren (X) (season 2007 only).

	Niesen Kulm (K)				Niesen Schwandegg (S)				Lägeren (L)				Lägeren (X)			
Ion	Min	Max	Median	Mean[a]	Min	Max	Median	Mean[a]	Min	Max	Median	Mean[a]	Min	Max	Median	Mean[a]
F^-	0.0	51.2	2.4	1.6	0.0	11.2	0.8	1.5	0.0	5.7	1.9	2.7	0.0	2.1	0.3	0.4
Cl^-	5.0	131.3	23.6	10.6	7.2	62.9	24.1	20.5	13.5	126.2	42.8	44.9	15.4	93.9	36.6	32.8
NO_2^-	0.0	5.4	1.2	1.1	0.0	3.6	1.0	1.1	0.0	43.5	12.0	15.6	0.0	20.6	3.2	5.4
NO_3^-	0.0	929.0	130.1	87.0	40.5	959.1	104.5	200.6	10.4	1816.4	505.1	347.7	7.7	668.9	272.1	294.3
PO_4^{3-}	0.0	122.5	9.8	6.9	0.0	284.7	20.7	13.9	0.0	1489.6	28.0	215.6	5.0	469.9	131.1	169.5
SO_4^{2-}	17.7	707.9	107.7	72.3	31.8	350.5	82.3	105.0	192.6	893.8	289.2	334.0	108.3	430.2	351.1	297.4
Na^+	3.1	272.3	53.5	43.0	5.2	141.3	57.0	44.3	12.6	223.9	55.2	78.0	0.0	111.7	52.5	38.2
NH_4^+	41.9	906.4	181.4	143.5	78.3	829.0	178.0	249.8	266.7	2128.8	517.5	1032.3	259.3	3374.3	854.9	844.5
K^+	2.9	83.9	8.3	5.0	5.5	166.0	12.8	10.3	8.4	449.6	26.2	96.1	6.0	619.0	80.4	72.0
Mg^{2+}	1.0	230.5	15.0	12.6	6.4	56.3	14.3	13.9	3.6	730.0	38.1	139.8	10.3	475.3	42.6	54.1
Ca^{2+}	13.2	3528.9	74.7	46.8	8.9	188.7	27.2	35.4	40.0	818.1	103.1	117.9	14.4	73.2	43.7	32.8
H^+	0.0	1.1	0.1	0.5	0.0	1.6	0.1	0.4	0.0	2.5	0.0	0.3	0.0	0.1	0.0	0.1
Lf	14.1	546.0	49.0	34.4	23.1	210.0	48.9	60.4	69.4	471.0	168.6	208.7	138.2	809.0	227.0	255.6
pH	6.0	7.7	6.8	6.4	5.8	7.6	6.8	6.6	5.6	7.5	7.3	7.0	7.0	8.1	7.3	7.2

[a] Volume weighted mean

Table 4: Ion concentrations [μeq l^{-1}], pH and conductivity [μS cm^{-1}] in fogwater and precipitation, and their ratios between the sites Niesen Kulm (K), Niesen Schwandegg (S), Lägeren L and X, and Niesen Kulm Precipitation (R) during the warm seasons 2006 and 2007. Asterisks denote a significant difference in volume-weighted means at a level of $p < 0.05$ (**) and $p < 0.10$ (*), respectively.

	Volume-weighted means					Ratios				
Ion	K	S	L	X	R	K/S	K/L	S/L	L/X	K/R
F^-	1.6	1.5	2.7	0.4	1.2	1.05*	0.61	0.58	3.84*	0.92*
Cl^-	10.6	20.5	44.9	32.8	8.2	0.52	0.24**	0.46**	0.96	0.88**
NO_2^-	1.1	1.1	15.6	5.4	0.2	1.00	0.07**	0.07**	0.88	5.43**
NO_3^-	87.0	200.6	347.7	294.3	10.9	0.43	0.25**	0.58**	1.27	6.76**
PO_4^{3-}	6.9	13.9	215.6	169.5	0.2	0.49	0.03	0.06	0.11**	11.13**
SO_4^{2-}	72.3	105.0	334.0	297.4	13.2	0.69	0.22**	0.31**	0.85	4.91**
Na^+	43.0	44.3	78.0	38.2	44.6	0.97	0.55	0.57	1.21	0.90
NH_4^+	143.5	249.8	1032.3	844.5	15.8	0.57	0.14**	0.24**	0.50	8.76**
K^+	5.0	10.3	96.1	72.0	4.4	0.48*	0.05**	0.11**	0.24*	0.83**
Mg^{2+}	12.6	13.9	139.8	54.1	8.8	0.90	0.09	0.10**	0.30	1.17**
Ca^{2+}	46.8	35.4	117.9	32.8	11.7	1.32**	0.40	0.30**	2.87**	2.23**
H^+	0.5	0.4	0.3	0.1	1.1	1.23	1.61	1.31	7.52	0.44
Lf	34.4	60.4	208.7	255.6	8.5	0.57	0.16**	0.29**	0.45	3.54**
pH	6.4	6.6	7.0	7.2	6.5	0.98	0.92	0.94	0.94	0.99

Table 5: Ion concentrations [μeq l^{-1}], pH and conductivity [μS cm^{-1}] of precipitation at the Niesen Kulm site for the season 2007.

Ion	Min	Max	Median	Mean[a]
F^-	0.0	3.9	0.4	1.2
Cl^-	1.2	30.9	2.5	8.2
NO_2^-	0.1	0.4	0.2	0.2
NO_3^-	3.8	22.7	13.7	10.9
PO_4^{3-}	0.0	0.4	0.1	0.2
SO_4^{2-}	3.4	22.7	12.7	13.2
Na^+	0.0	64.2	40.8	44.6
NH_4^+	0.0	37.2	19.8	15.8
K^+	1.5	11.7	2.3	4.4
Mg^{2+}	0.0	10.2	9.2	8.8
Ca^{2+}	6.1	101.0	10.9	11.7
H^+	0.0	7.9	0.4	1.1
Lf	4.3	21.3	8.0	8.5
pH	5.1	7.7	6.3	6.5

[a] Volume weighted mean

Table 6: Comparison of weighted mean concentrations of selected ions (μeq l^{-1}) and pH from measurements conducted during this study and selected results from other studies (based on a compilation by Burkard et al. 2003).

	$NO_3^- + NH_4^+$	NO_3^-	NH_4^+	SO_4^{2-}	pH	Cl^-	Ca^{2+}	Mg^{2+}	K^+	Na^+	Reference
Fog Under Advective Influence (FAI)											
Mount Rigi, Switzerland	1620	520	1100	430	5.2	8	98	24	16	18	Collett et al. [1993b]
Waldstein, Germany	1390	464	926	438	4.1	43	54	13	12	47	Thalmann et al. [2002]
Lägeren	1361	623	738	338	4.6	50	69	8	33	84	Burkard et al. [2003]
Mont Tremblant, Québec	409	170	239	339	3.7	13	70	23	6	10	Schemenauer et al. [1995]
Radiation Fog (RF)											
Mantova, Italy	7365	1594	5771	2724	6.6	360	163	33	40	37	Fuzzi et al. [1996]
Lägeren	2748	1200	1548	678	4.3	109	121	11	93	218	Burkard et al. [2003]
Lägeren	1955	547	1408	660	5.2	–	–	–	–	–	Joos and Baltensperger [1991]
San Joaquin Valley, California	1491	483	1008	117	6.5	15	9	–	–	–	Collett et al. [1999]
Further Studies											
Milesovka Observatory, Czech Republic	1602	524	1078	3022	4.6	85	545	221	32	90	Fisak et al. [2009]
Waldstein, Germany	1150	481	669	497	4.3	54	69	20	12	65	Wrzesinsky and Klemm [2000]
Flaje, Czech Republic	929	726	203	1250	3.0	155	136	40	19	64	Bridges et al. [2002]
Vallombrosa, Italy	665	301	364	468	3.6	–	–	–	–	–	Cini et al. [2002]
Mount Rokko, Kobe, Japan	565	301	264	313	3.8	230	121	72	15	247	Aikawa et al. [2005]
Szrenica, Poland	450	240	210	200	4.6	100	140	49	45	100	Błaś et al. [2010]
Whiteface Mountain, NY[a]	228	79	149	220	3.9	7	26	7	2	2	Aleksic et al. [2009]
This Study											
Lägeren (X)	1326	294	1032	297	7.2	33	33	54	72	38	
Lägeren (L)	1193	348	845	334	7.0	45	118	140	96	78	
Niesen Schwandegg (S)	451	201	250	105	6.6	21	35	14	10	44	
Niesen Kulm (K)	231	87	144	72	6.4	11	47	13	5	43	

[a] Data from 2006 only